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Investigation of H_2 Concentration and Combustion Instability Effects on the Kinetics of Strained Syngas Flames

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Preface

This is the final technical report on DOE Grant DE-FG26-05NT42495 entitled "Investigation of H_2 Concentration and Combustion Instability Effects on the Kinetics of Strained Syngas Flames". Technical work supported by this grant will be a part of Mr. Mahesh Subramanya's doctoral dissertation.

Abstract

The flame extinction limits of syngas (H_2-CO) flames were measured using a twin-flame-counter-flow burner. Plots of Extinction limits vs. global stretch rates were generated at different mixture compositions and an extrapolation method was used to calculate the flame extinction limit corresponding to an experimentally unattainable zero-stretch condition. The zero-stretch extinction limit of H_2-CO mixtures decreases (from rich to lean) with the increase in H_2 concentration in the mixture. The average difference between the measured flame extinction limit and the Le Chatelier's calculation is around ~ 7%. The measured OH^* chemiluminescent data indicates that regardless of mixture compositions the OH radical concentration reduces (within the experimental uncertainties) to an extinction value prior to the flame extinction. Flame extinction limits of H_2-CO mixtures measured in a flat-flame burner configuration also show a similar relation. Additionally, the measured laminar flame velocity close to the extinction indicates that regardless of fuel composition the premixed flame of hydrogen fuel blends extinguishes when the mixture laminar flame velocity falls below a critical value. The critical laminar flame velocity at extinction for H_2-CO premixed flames (measured in the flat flame burner configuration) is found to be 3.77(±0.38) cm/s. An externally perturbed H_2-CO twin flame was not experimentally achievable for the mixture conditions used in the present investigation. A slightest perturbation in the flow-field distorts the H_2-CO twin-flame. The flame becomes highly unstable with the introduction of an externally excited flow oscillation.

Executive Summary

Future generation gas turbine combustors are expected to utilize fuel compositions ranging from that of natural gas to a broad range of syngas without compromising low emission characteristics. To address the fuel variability and fuel flexibility issues of gas turbine combustors, fundamental information and data regarding the combustion characteristics of commercial and alternative fuels are essential. Despite this immense practical importance, little information is available regarding the combustion behavior of hydrogen containing synthesized fuel blends. The primary objective of the project is to systematically investigate flame extinction strain rates and flame structures of H_2-CO/air flames using a counterflow burner.

The present investigation used a twin flame counterflow burner to measure flame extinction and heat release processes of syngas flames at different H_2 concentration and stretch conditions. This burner uses a planar, twin-flame-counterflow nozzle system to determine the stretch rate at which flames are extinguished. By performing experiments at decreasing fuel/air ratios, it is possible to plot the equivalence ratio versus the extinction stretch rate, and to extrapolate the results to quantify the equivalence ratio corresponding to an experimentally unachievable zero-stretch condition. Chemiluminescent measurements of OH and CH radical concentrations were used to characterize the heat release processes of the flame.

The flame extinction limits of pure CH_4 [99.97%] was measured for the qualification and validation of the present twin flame counter flow burner system. The measured zero stretch extinction limit (in %f) of CH_4 at the standard laboratory condition is 4.66±0.12. The measured value is within 1% of the value reported in the literature. The zero-stretch extinction limit of H_2-CO mixtures decreases (from rich to lean) with the increase in H_2 concentration in the mixture. The average difference between the measured flame extinction limit and the Le Chatelier's calculation is around ~ 7%. The OH^* chemiluminescent data indicates that regardless of mixture compositions the OH radical concentration reduces (within the experimental uncertainties) to an extinction value prior to the flame extinction. The critical laminar flame velocity at extinction for H_2-CO premixed flames is found to be 3.77(±0.38) cm/s.

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Nomenclature

a	= empirical coefficients
K	= Global stretch-rate, U/y
U	= Exit velocity of air-fuel mixture
y	= Distance between stagnation plane and nozzle exit
%f	= volumetric percent-fuel in air-fuel mixture
f_0	= extinction value of base fuel (in percent fuel)
$\%f_{ext}$	= percent-fuel in air-fuel mixture at extinction
x_{H2}	= volumetric composition of Hydrogen in fuel blend

I. Introduction

With an emerging need to address gas turbine combustor issues such as fuel variability and fuel flexibility, fundamental information and data regarding the combustion characteristics of commercial and alternative fuels are essential [**1-2**]. Future generation gas turbine combustors are expected to tolerate fuel compositions ranging from that of natural gas to a broad range of syngases without sacrificing low emission characteristics [**3**]. Hydrogen containing fuel blends such as various coal synthesized gases (commonly known as syngas) have unique flame dynamics and kinetic behaviors [**4-6**]. Even a small amount of hydrogen in a fuel can trigger the onset of flashback by altering the kinetics and transport properties of the fuel mixture. Thus, the presence of hydrogen in the fuel mixture significantly changes the flame response to stretch, preferential diffusion and instabilities. Despite this immense practical importance, little information is available regarding the combustion behavior of hydrogen containing synthesized fuel blends. Although it may be expected that the combustion characteristics of fuel blends will fall between those of the primary fuels, some recent investigations have clearly demonstrated that blended fuels can have significantly different characteristics from those of the original fuels [**4-6**].

From the fundamental view point, the information regarding the extinction behavior of a particular fuel mixture is important to understand the basic combustion mechanisms. The term *flame extinction limit* has a different implication compared to the general

definition of *flammability limit*. Generally flammability limit is defined as an experimentally determined limiting concentration of fuel in an oxidizer beyond which combustion does not to occur in a standard apparatus and at a set of standard test conditions. Hence, the experimental evaluation of such limits is dependent on apparatus and test protocol. In contrast, flame extinction limit is defined as a certain limit mixture of fuel and oxidizer in which propagation of a steady one-dimensional premixed flame is not possible [**7-8**], and is a more fundamental thermo-chemical characteristic. Various parameters such as fuel type, mixture properties and mass diffusion are some factors in defining the limiting compositions. From the theoretical perspective, flame extinction occurs since energy loss rate due to mechanisms such as chain-terminating reactions, radiative heat loss, and non-equi-diffusion eventually exceeds the energy release rate and causes the extinction of the flame at the limit concentration.

In recent years there is a renowned interest of developing kinetic mechanism for hydrogen fuel-blends [especially H_2-CO mixtures) due to a rapid increase in use of hydrogen fuel blends [coal blown syngas] in terrestrial gas turbine based power generations [**1-2**]. Additionally, dynamics of CO flame doped with small amount of H_2 serves as an excellent platform for kinetic studies. Since the primary kinetics of CO_2-H_2 are dominated by well studied CO+OH = CO_2+H reaction, accurate experimental measurements of CO-H_2 flame characteristics can be reliably used to benchmark kinetic mechanisms of fuel blends [**9**]. Unlike flammability limit data, which are generally tied to a specific experimental condition, flame extinction measurements are highly

amenable to computational verifications. Previously Maruta et al., 1996 [**10**], Guo et al., 1997 [**11**] have shown a good comparisons of computed and measured flame extinction limits of CH_4-air flame for normal and microgravity conditions. However, currently combustion literature lack flame extinction data for most of the hydrogen fuel blends. This report intends to provide accurate extinction data for H_2-CO mixtures. The range of mixture conditions used in the present investigation represents typical mixture conditions of syngas (up to 30% H_2 in H_2-CO mixture). Additionally, beyond this range (greater than 30% H_2) flame extinction measurements using a twin-flame configuration are not experimentally achievable due to severe flashback problems.

The most widely accepted technique for determining the *flame extinction limit* utilizes a twin flame counterflow arrangement proposed by Ishizuka and Law, 1982. [7]. This technique uses a planar, twin-flame-counterflow nozzle system to determine the stretch rate at which flames are extinguished. By repeating the experiment at diminishing fuel/air ratios, it is possible to plot the equivalence ratio versus the extinction stretch rate, and to extrapolate the results to identify the equivalence ratio corresponding to an experimentally unattainable zero-stretch condition. Unlike the ignition method for the flammability limit measurement, this technique entirely avoids issues associated with the ignition system.

Twin flame counter flow configuration allows quantifying the effects of hydrodynamic strain on flames. This is essential to understand how turbulence induced strained affects

flame behavior (such as extinction and flashback) in practical combustors. In this configuration unburned reactions are issued from two identical burners placed in opposed flow configurations and twin flames are formed on both side of the stagnation flame. It can be shown* that the global flame stretch, K, is the magnitude of the axial velocity gradient U/z [**12**]**.**

A common relation which can be used to evaluate the flammability limit of fuel blends is known as Le Chatelier's Rule†. The rule uses an additive formula to calculate the flammability from the limiting mole fractions X_i of each species 'i'. *However, prior to the current study, applicability of the rule has never been experimentally verified to estimate the flame extinction limit*. The extinction behavior of a fuel largely depends on complex non-linear interactions of burning velocity, energy release and external loss mechanisms. When multiple fuels are present in a mixture, the burning velocity and energy loss mechanisms of the fuel blend may have different relation with the mole fractions of the each individual fuel. In fact, previously several investigators have noticed that in a fuel blend, the fuel with the lower burning velocity generally acts as a flame inhibitor [**14**] and slows down the reaction processes. Additionally, the fuel blends have different

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* Quote "The axial flow velocity, U, is given by ±2cz, where c is constant and z is the axial distance from the stagnation plane, which is independent of the radial coordinate r. The radial velocity, V, is given by cr and is independent of z. The stretch acting on the flame is the sum of the extensional strains in the two directions orthogonal to the z axis, that is, $\frac{\partial V}{\partial x}+\frac{\partial V}{\partial y}=2c=-\frac{\partial U}{\partial z}$. Thus the flame stretch Σ is simply the magnitude of the axial velocity gradient." Unquote [**12]**

† Quote " $L=\frac{100}{\frac{P_1}{L_1}+\frac{P2}{L2}+\frac{P_3}{L3}+\cdots\cdots}$ in which L is the lower limit (volume percentage) of a complex fuel gas mixture and P_1, P_2, P_3 ….. are the proportions (percentage) of each fuel gas in the mixture, free from air and inerts, so that $P_1 + P_2 + P_3 + \ldots = 100$, and $L_1+L_2+L_3 + \ldots$ are the lower flammability limits of the individual components." Unquote. [**13**]

physico-chemical characteristics compared to their pure counter parts and it is still unknown how these shifts in physico-chemical properties will impact burning velocity and preferential diffusion. Thus, the prediction accuracy of Le Chatelier's relation for determining the extinction limit of fuel mixtures needs to be quantified.

Since previous investigations [**12**] have indicated that the extinction behavior is weakly affected by the rate parameters, extinction limit of fuel blends may be inferred by comparing measurements of two different experimental configurations (twin-flame and flat flame) without detailed chemical knowledge. In addition, instead of using limit compositions, a better comparison can be drawn by comparing laminar flame velocity (S_L) of different fuel mixtures at the limit. Consequently, the present investigation deals with the flame extinction data of hydrogen fuel blends measured at different compositions and experimental configurations. A Twin Flame Counter flow burner and a flat flame burner were used to measure the lean flame extinction limits. The flame extinction behaviors are analyzed in the context of flame velocity at the extinction limit. Flame temperature and chemiluminescent emission measurements of OH^* and CH^* were also used to gain more understanding of the flame velocity of hydrogen fuel blends. In this paper, instead of equivalence ratio (ϕ), flame extinction limit is expressed as a volumetric percent of fuel (%f) in a fuel-air mixture. This is done to avoid defining mixture equivalence ratios for fuel blends which is often confusing and ambiguous since different fuels have different stoichiometric air requirements.

In the context of above discussion the report is first to provide the zero-stretch flame extinction limits of H_2-CO mixtures. As stated earlier the data will be particularly useful for modeling and design purposes of fuel flexible gas turbine combustors.

Periodic perturbation on the flow-field affects the flame dynamics in several ways. It is important to understand the response of the strained lean flames to periodic flow oscillation due to its relevance to combustion instabilities and practical turbulent flames [**15-16**]**.** Previously investigators have suggested that the flame responds distinctly with the change in oscillation frequency and have identified a low and a high frequency limit [**15**]. The flame response to the excitation frequency is 'quasi-steady' for an excitation frequency below the lower limit. The Flame response time is shorter in compared to the oscillation time scale when the lean flame is subjected to an excited frequency smaller than the lower limit. In the event of an excitation frequency larger than the upper limit the flame response is longer in compared to the excitation time scale. This is due to the fact that flame properties cannot adjust to the rapid change in flow-field at a high frequency external oscillation and assume 'mean values' [**15**]. In between frequencies the flame extinction limit [the original literature defined it as a 'dynamic flammability limit'] decreases [become leaner] with the introduction of an externally excited periodic flow oscillation [**16**]. However, the coupling synergism of acoustic perturbation and multi component fuel compositions is yet to be studied. The present study advance the understanding of the response of the multicomponent fuel to the effects of stretch and acoustic disturbances.

II. Experimental Techniques

This chapter discusses the design of counter-flow twin-flame burner, construction of fuel-air train and various data acquisition systems used, and the methodology practiced in evaluating the flammability limits.

2.1 Counter-flow Twin-flame Burner Assembly

The counter-flow twin-flame burner assembly is engineered to control the combustion conditions and achieve flame extinction processes over a wide flame-stretch range. In most applications, flame stretch is controlled by varying air and fuels' flow rates. The burner assembly in the current study is capable of varying the top burner's position (i.e., the distance between top and bottom burner exits), thereby providing a wider range and control over flame stretches. The top burner is mounted on a computer-controlled high-precision traverse mechanism facilitating its position over the identical bottom burner on the same vertical axis. The two identical burner-assemblies consist of five distinctive parts each, namely; acoustic housing, manifold, stem, connector and burner nozzle exit. To optimize weight of the burner assemblies, especially on the traverse mechanism, acoustic source housings, manifolds, stems and connectors are fabricated with aluminum. The burner nozzles are fabricated with brass, selected for its better thermal properties over aluminum. **Figures 2.1a-b** show the construction of the burner assembly.

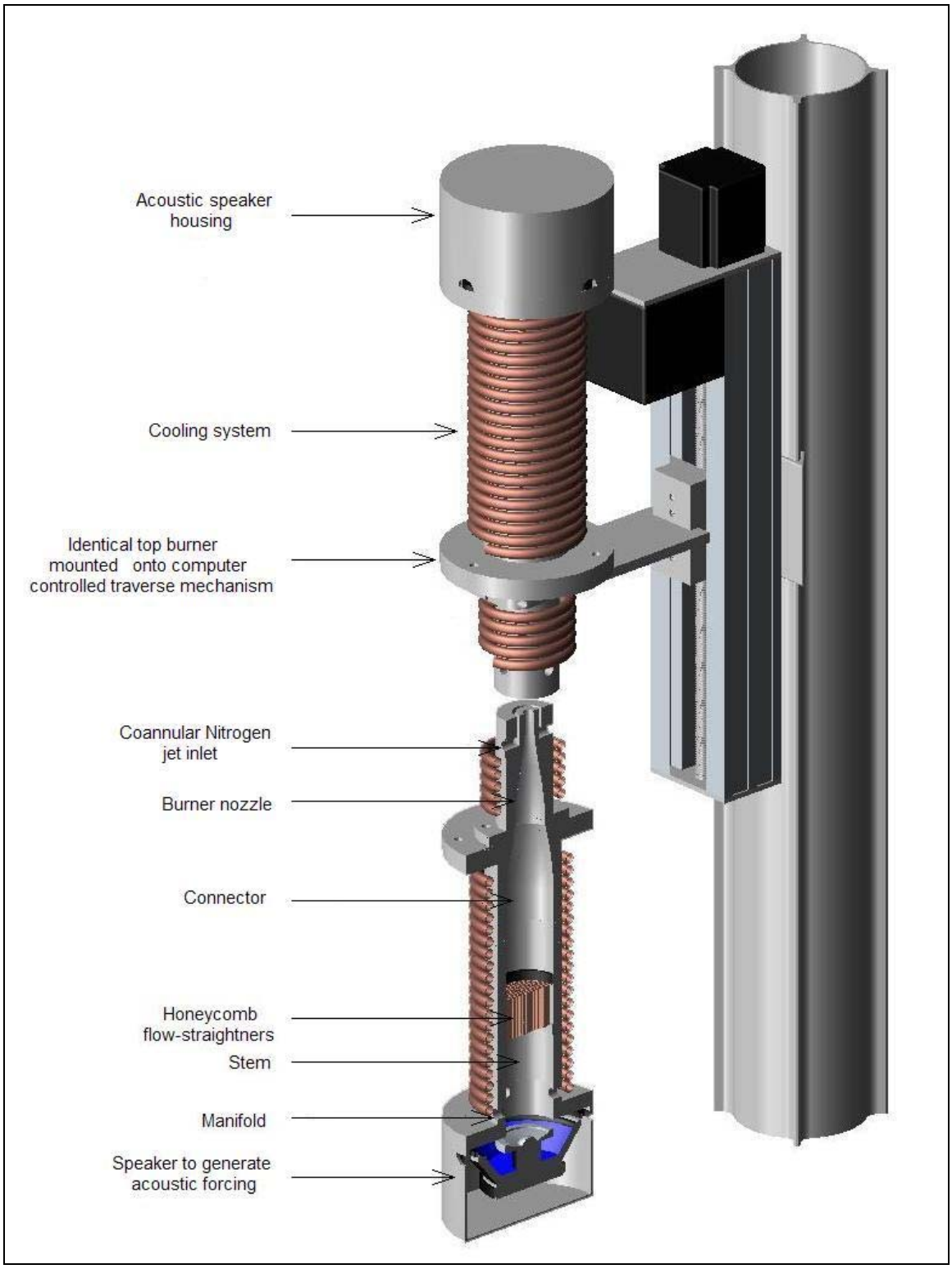

Figure 2.1a: Counter-flow twin-flame burner assembly

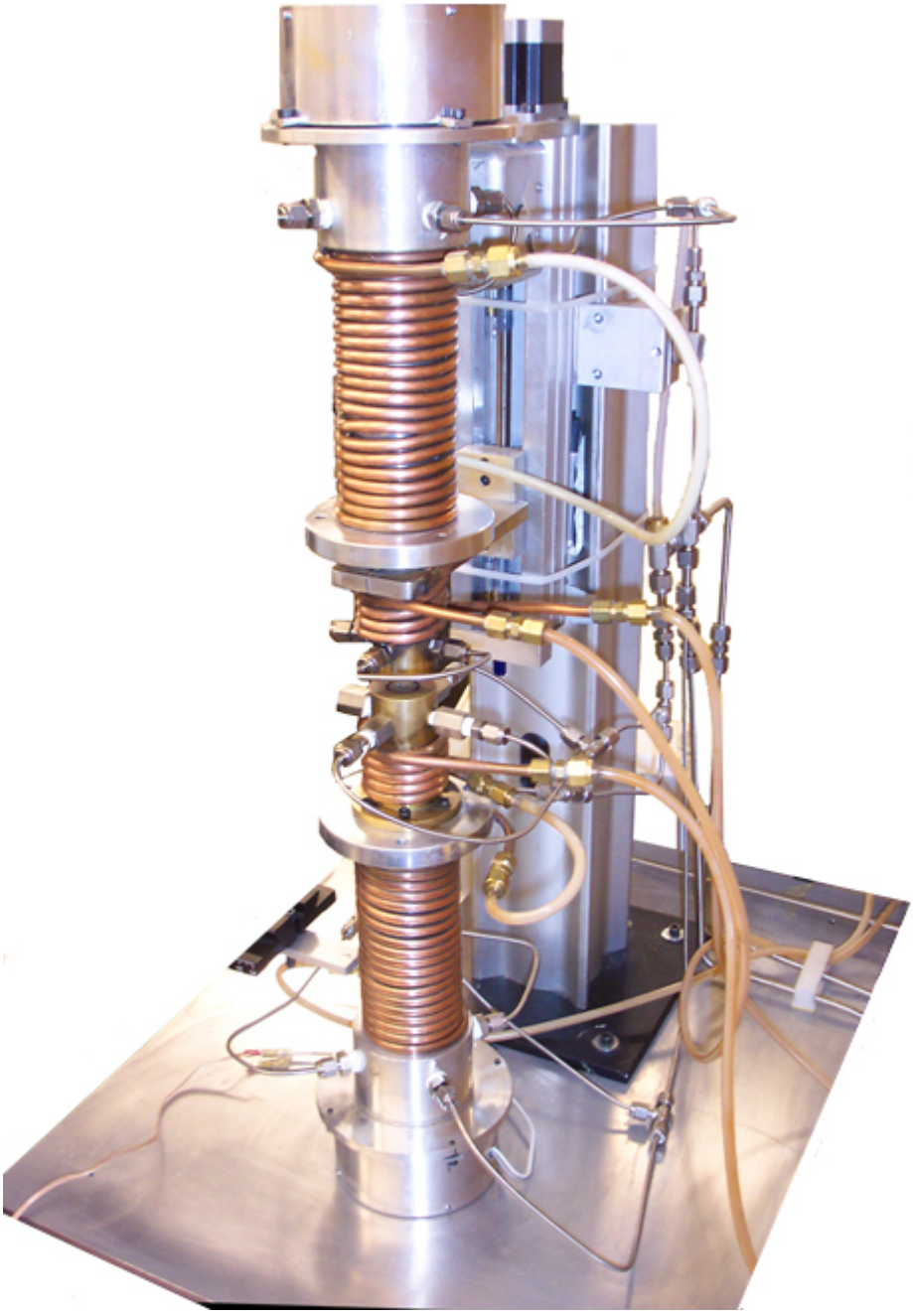

Figure 2.1b: Photograph of the Counter-flow twin-flame burner assembly

The acoustic source housing is designed to create an external excitation on the twin-flames and analyze the effect of acoustic oscillations on flame extinction limits. The flow

excitation is generated by a thin copper membrane vibration source fed by a 100W speaker. Speaker signal strengths (in terms of their amplitudes and frequencies) are controlled through an amplifier (AudioSource Amp 100 amplifier) and a signal synthesizer (TrueRTA software). Using a thermal anemometer, the effect of these acoustic forcing (100Hz, 300Hz, 500Hz and 700Hz forcing frequencies) on velocities and turbulence-intensities of flow-field at burner exits are measured and shown in **Figures 2.2 and 2.3**. **Figure 2.4** shows the microphone, speakers, amplifier and system used for signal synthesizer software. The thermal anemometer and its working principle are discussed later in the chapter.

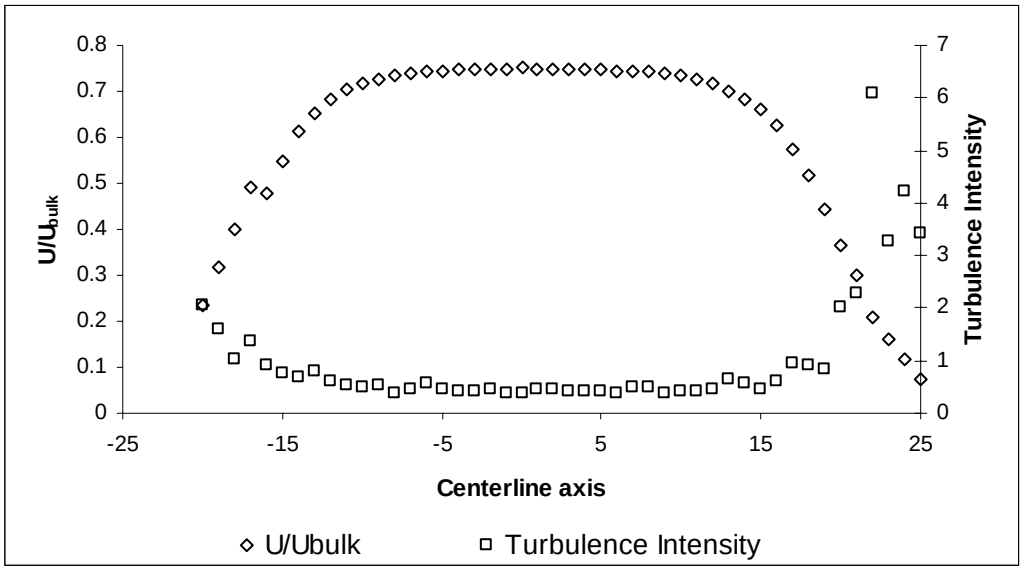

Figure 2.2: Hotwire Anemometry data on the burner exit profile analyzed without the effect of acoustic oscillations

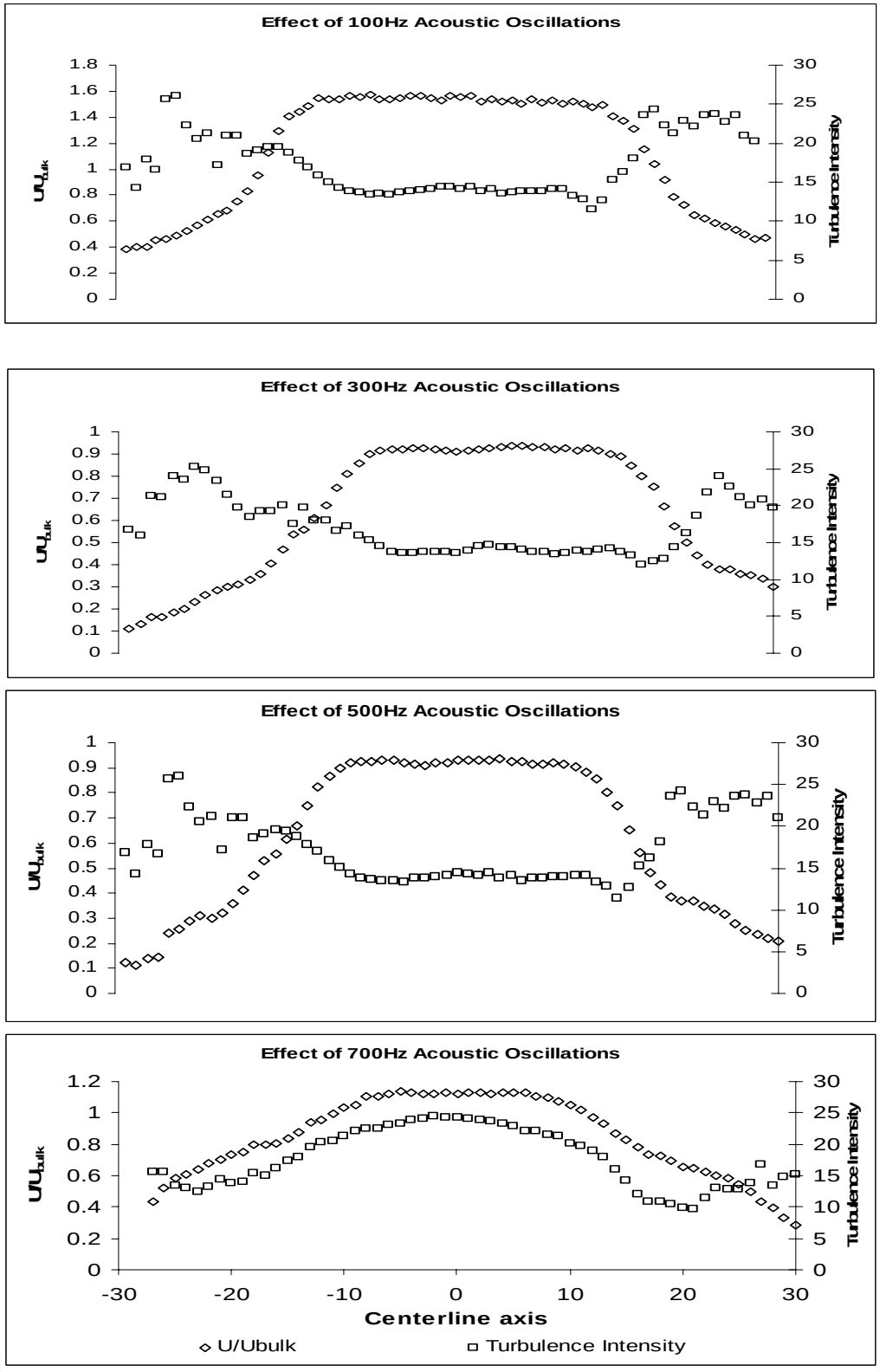

Figure 2.3: Hotwire Anemometry data on the effect of acoustic oscillations on burner exit velocity

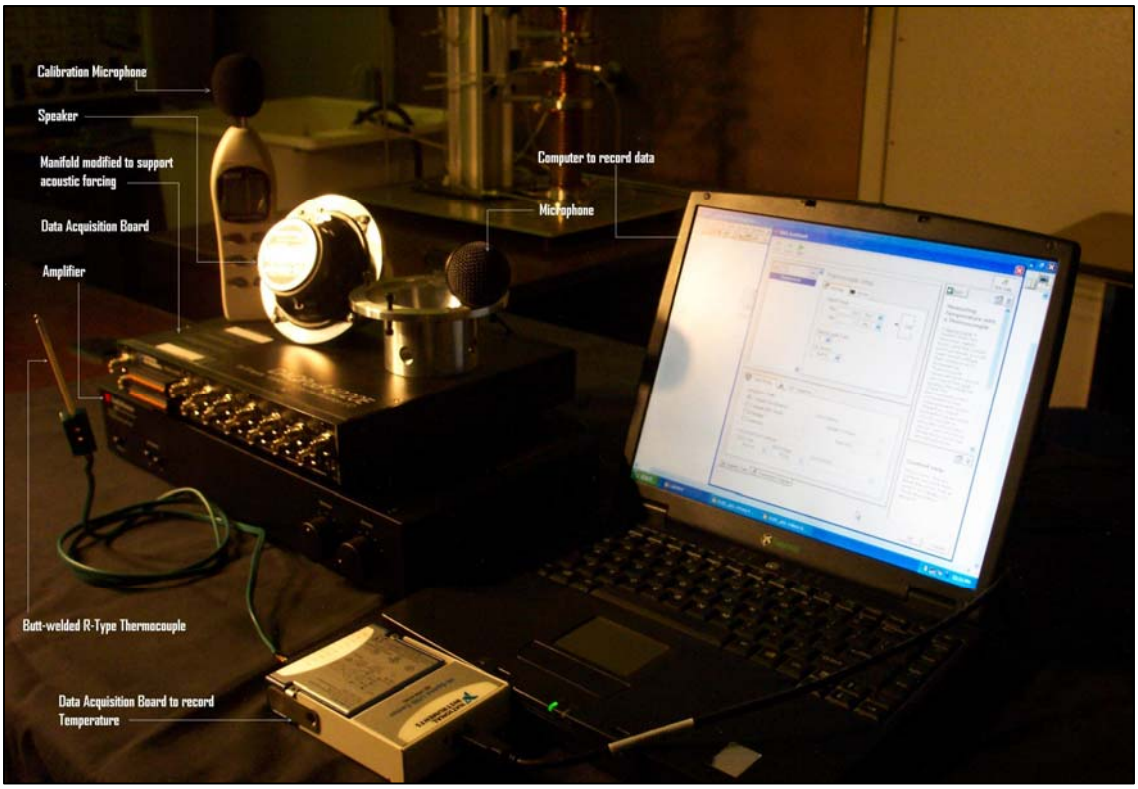

Figure 2.4: Acoustic forcing generation and measurement system

Air and fuels' (single and blended) mixtures from air-fuel train are fed to the top and bottom burner assemblies through four symmetric insertion inlets on the manifolds. Since stratified fuel effects can have an adverse effect on the flammability limit analysis, homogenous mixing of air and fuels are ensured by blending the streams in T-connectors positioned upstream to manifolds. With one end of the manifold connected to acoustic source housing, the other end leads to the stem of the burners. Injection induced eddies are filtered by wire-meshes mounted in the stems. The air-fuel streams are then passed through honeycomb matrices that function as a natural flow-straightner. Laminar air-fuel flow streams from the stems are lead through the burner connectors engineered to reduce the conditioned air-fuel stream diameter to enter the burner nozzle.

The burner nozzles are designed to provide suitable exit conditions such as uniform exit flow velocity and to shield the twin-flames from ambience. As seen in Figure 2.5, the nozzle curve from inlet to exit follows a fifth-order polynomial relation, expressed as in Equation 2.1.

$$r(\ell) = -0.00925925\ell^5 + 0.0694444\,\ell^4 - 0.138888\ell^3 + 0.5 \quad \text{... Eq. 2.1}$$

This polynomial equation was developed through a CFD analysis [**17**] to insure uniform exit velocity. Derivation of the nozzle curve's polynomial trend, with the boundary conditions considered, can be found elsewhere [**17**]. A Constant Temperature Hot-wire Anemometry analysis of the flow exit conditions, shown in **Figure 2.2**, validates the uniform exit flow velocity with minimal wall-effects. Low turbulence intensity measurements observed from the same analysis validates the construction of flow conditioners upstream in the burner stems. A Nitrogen flow passage is constructed concentric to the burner nozzle exit and serves multiple purposes. Firstly, the nitrogen co-flow protects the twin-flame from the influence of surrounding air. This insures the equivalence ratio of flames in the combustion zone remains controlled until flame extinction is witnessed. Secondly, the nitrogen co-flow furthermore trims any wall-effects present in the flow exit. Thirdly, the high-temperatures formed at the burner tip are significantly cooled with the nitrogen co-flow.

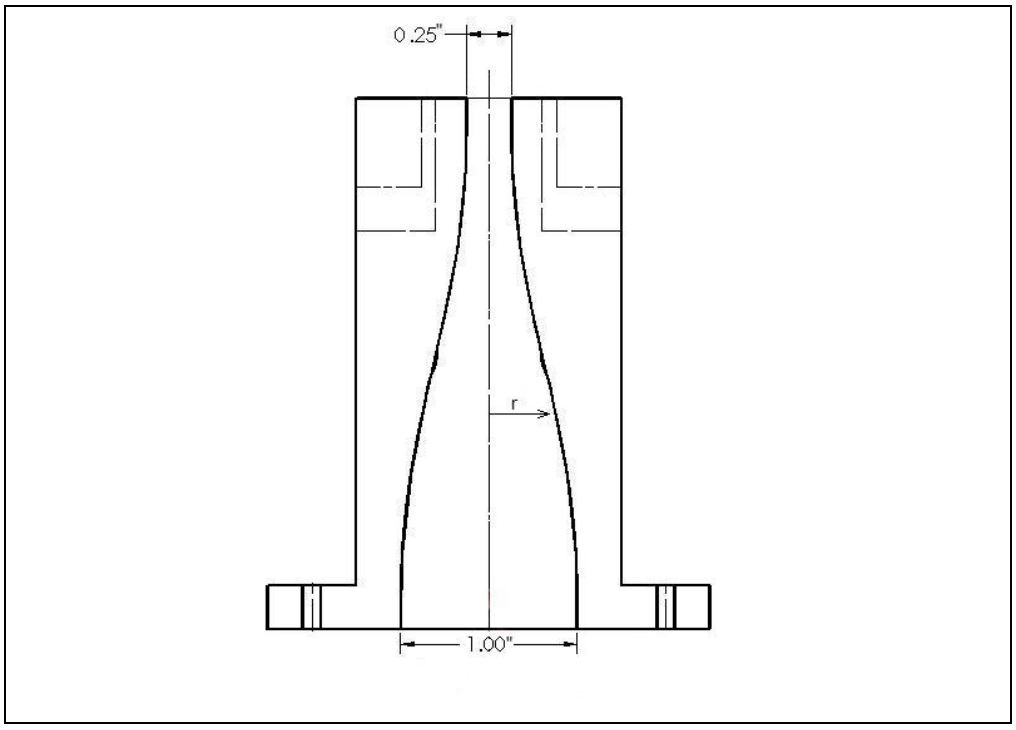

Figure 2.5: Fifth-order polynomial curve reducing the nozzle inlet from one-inch to outlet 0.25-inches

Pre-heating of air-fuel mixtures upstream can significantly lower the lean flammability limits of fuels. Therefore, the burner assemblies are wound with copper coils and are continuously circulated with cold-water to insure that the air-fuel mixtures enter combustion zone at room-temperature. As discussed earlier, the nitrogen co-flow also reduces the high-temperatures due to flames at the burner tip significantly.

2.2 Air-Fuel Flow Train

Individual lines, each with a digital mass flow-meter, precision metering valve and a low-torque-quarter-turn plug valve, are used to control and quantify air and fuels flows. T-connectors are used upstream to the burner assemblies to mix the fuels and air, and to

ensure homogenous supply of air-fuel mixture to the combustion zone. In addition, the flow system is designed such that it avoids any stratified effect of fuels to dominate during combustion. Since premixed combustion processes are prone to flashbacks, especially when using hydrogen blend fuel mixtures, a 2-micron flame arrestor is mounted on the air-fuel train. The flow to bottom burner assembly is controlled by an additional metering valve, so as to compensate for the pressure losses towards the top burner and bifurcate equally the air-fuel mixtures between top and bottom burner assemblies. Equal quantities of air-fuel mixture flows to top and bottom burners are confirmed by the position of twin-flames (and stagnation plane) mid-way between the two burners.

2.3 Traverse Mechanism

The distance between the top and bottom burners is controlled by the position of the top burner assembly mounted on a computer controlled high-precision traverse mechanism. Since flame-stretch in one-dimensional flames is a ratio of air-fuel stream exit-velocity and the distance between the burner exit and stagnation plane, the adjustable positioning of top burner facilitates the test-matrix to cover a wide range of flame-stretches of various fuel compositions and conditions.

2.4 Image Acquisition System

A Charge-Coupled-Device camera is used to capture magnified, real-time images of the twin-flames' extinction process. These images aid in the judgment of the twin flame's

alignment and also provide a better insight to the flame extinction process. For instance, the effect of diffusional demixing, also called as Lewis Number effect, is well observed in magnified images that will otherwise be difficult to witness. Lewis Number, ratio of thermal diffusivity to mass diffusivity of gases in mixtures, is greater for fuels with lower carbon atoms such as methane in comparison to fuels with greater carbon chains such as propane. Therefore, during the flame extinction process of methane-methane twin-flames, the flames appear closer to stagnation plane as it stretches towards extinction

2.5 Thermal Anemometry

A constant temperature anemometry system is used in characterization of air flow-field at burner exits [**Figure 2.6**]. The thermal anemometer measures velocity of the flow-field by sensing the changes in heat transfer from its sensor which is a small, electrically-heated wire, of diameter four to six microns, exposed to the air or nitrogen passage exiting the burner. Reduce in sensor temperature due to the air stream is balanced by current to maintain a constant temperature, and the magnitude of this current is calibrated to measure the velocity of the stream. Figures 2.2 and 2.3 presented earlier to discuss the effect of acoustic forcing on the flow-streams show the velocity and turbulent-intensity readings measured using thermal anemometry at different conditions.

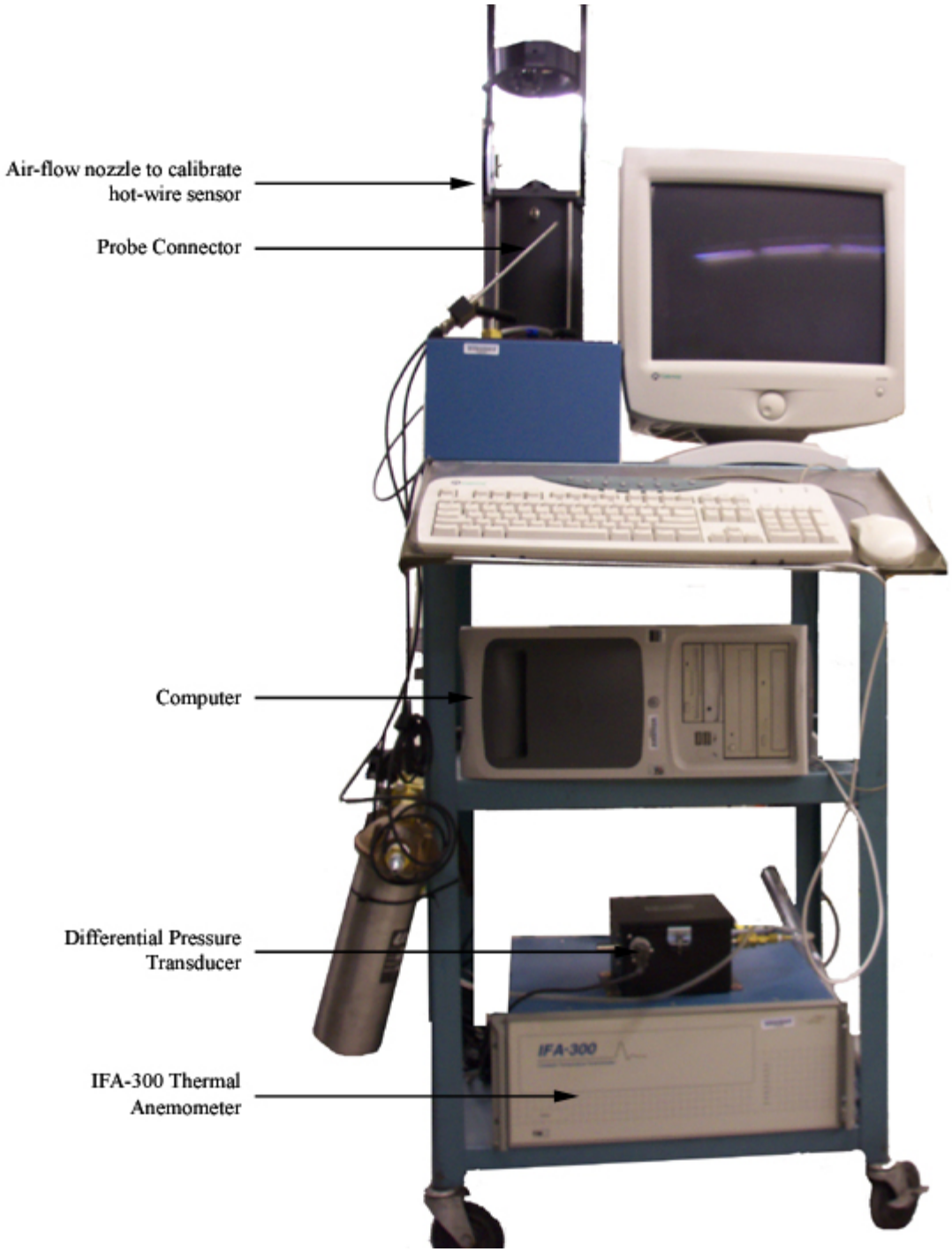

Figure 2.6: Thermal anemometry system (Inset: Hot-wire sensor of diameter four to six microns)

2.6 Spectroscopy

A computer controlled triple grating monochromator is used to measure chemiluminescent emissions from OH and CH radicals [**Figure 2.7]**. A pair of plano-convex lenses is used to focus the flame emission on the input slit of the monochromator. A 500 microns pinhole mounted on the input slit is used to define the probe volume. The light intensity at the desired wavelength was monitored by a high sensitivity photomultiplier tube (PMT). The photomultiplier output is amplified and acquired through a National Instruments DAQ system. The output of the PMT at a particular wavelength was sampled at 1 kHz rate and online averaged over 5 seconds.

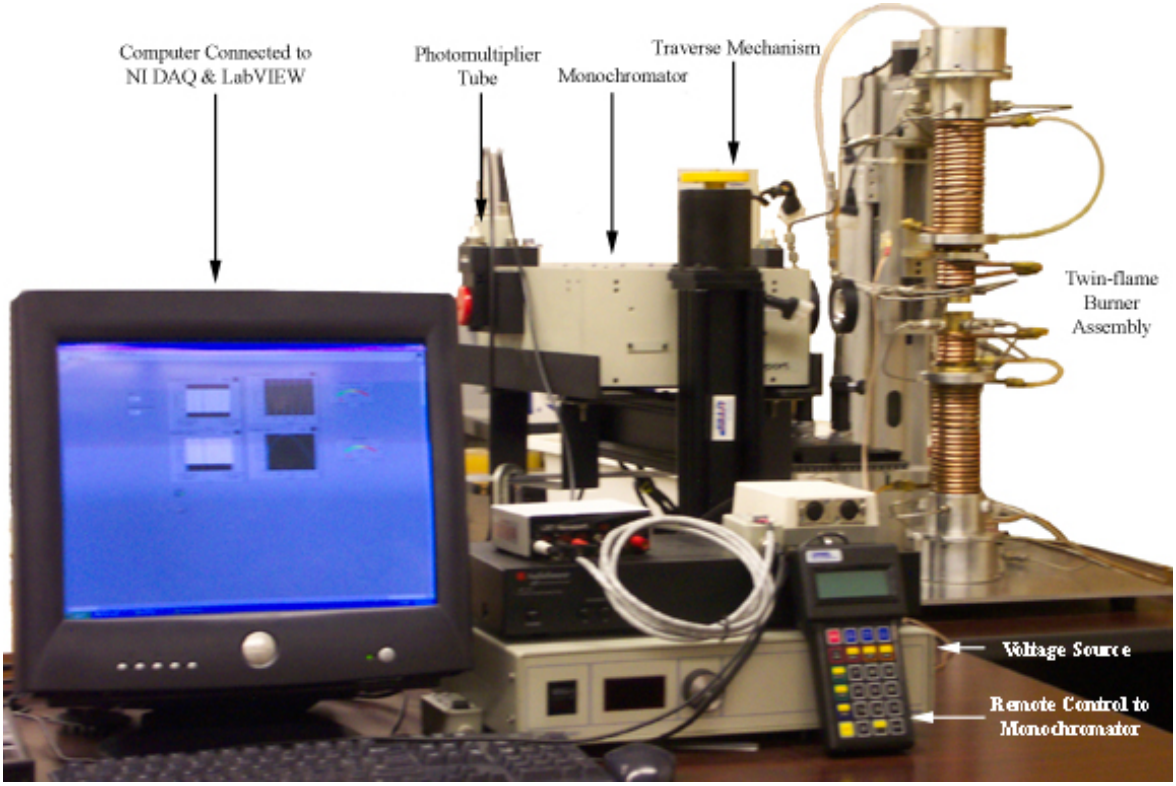

Figure 2.7: Computer controlled triple grating monochromator system

2.7 Methodology

Pre-determined fuel flow-rates/fuel blend compositions are fed through the burner system and the homogenous fuel mixture at burner exit is ignited with a pilot flame. Nitrogen is introduced through the concentric annulus of burner exit isolating the diffusion-twin-flames from atmospheric air, and a metered flow-rate of air is gradually added to the fuel mixture. As the air flow-rate is increased in fuel-air mixture, increase in flame-stretch is observed which eventually leads the twin-flames to extinction. Equivalence ratio and percent-fuel in air-fuel mixture at this flame-extinction condition is calculated. The process is repeated by varying the fuel flow-rates and maintaining the same mixture composition, thereby providing flame-stretch values at extinction for equivalence ratios ranging from 0.9 through the least experimentally attainable condition. Although the physio-chemical characteristics of fuels during combustion vary at very lean and very rich conditions, the characteristics are assumed to be similar to characteristics obtained as in earlier conditions due to experimental limitations. Therefore, the percent-fuel in air-fuel mixture is linearly extrapolated to zero-stretch conditions and the obtained percent-fuel at zero-stretch indicates the Lower Flammability Limit of fuel composition. Likewise, the experimental matrix is extended to varying fuel blend compositions to understand their effect on flammability limits.

2.8 Flat flame Burner

The flat flame burner system consists of fuel-air supply train with flow monitoring equipment, image acquisition, temperature measurement and water cooling systems.

Figure 2.8 shows the flat flame burner apparatus consisting of a 6 cm diameter porous metal disk burner. It is constructed from stainless steel and consists of a burner tube on the top of which rests the 6cm diameter disk. A 0.5 cm wide porous strip is concentric to the burner exit from which nitrogen passes to create an inert environment around the flame. The water cooling system extends throughout the entire porous disk system entering at the base of the burner. Water flow rate could be adjusted to vary the heat loss rate from the burner. The fuel/air mixture enters through the burner tube and passes through the porous media creating a uniform flow at the top of the disk.

The flames were ignited at fuel-rich mixture conditions and subsequently adjusted to their desired flow rate using precision needle valves. Once a stable flame front was obtained the nitrogen flow rate was adjusted so as to truncate the outlying edges. For extinction limit measurements the equivalence ratio was reduced while maintaining the fuel composition until the flame extinction occurs. The process was repeated for several heat loss rates from the burner and then extrapolated to zero heat-flux conditions to measure the flame extinction at the adiabatic condition. Temperature measurements were obtained with the use of an R -type thermocouple, mounted on a precision traverse mechanism placed at the burner surface to a set distance above the surface. Qualitative OH radical images were also captured using the Planar Laser Induced Fluorescence (PLIF) technique to understand the effects of hydrogen concentration on the flame velocity of fuel blends.

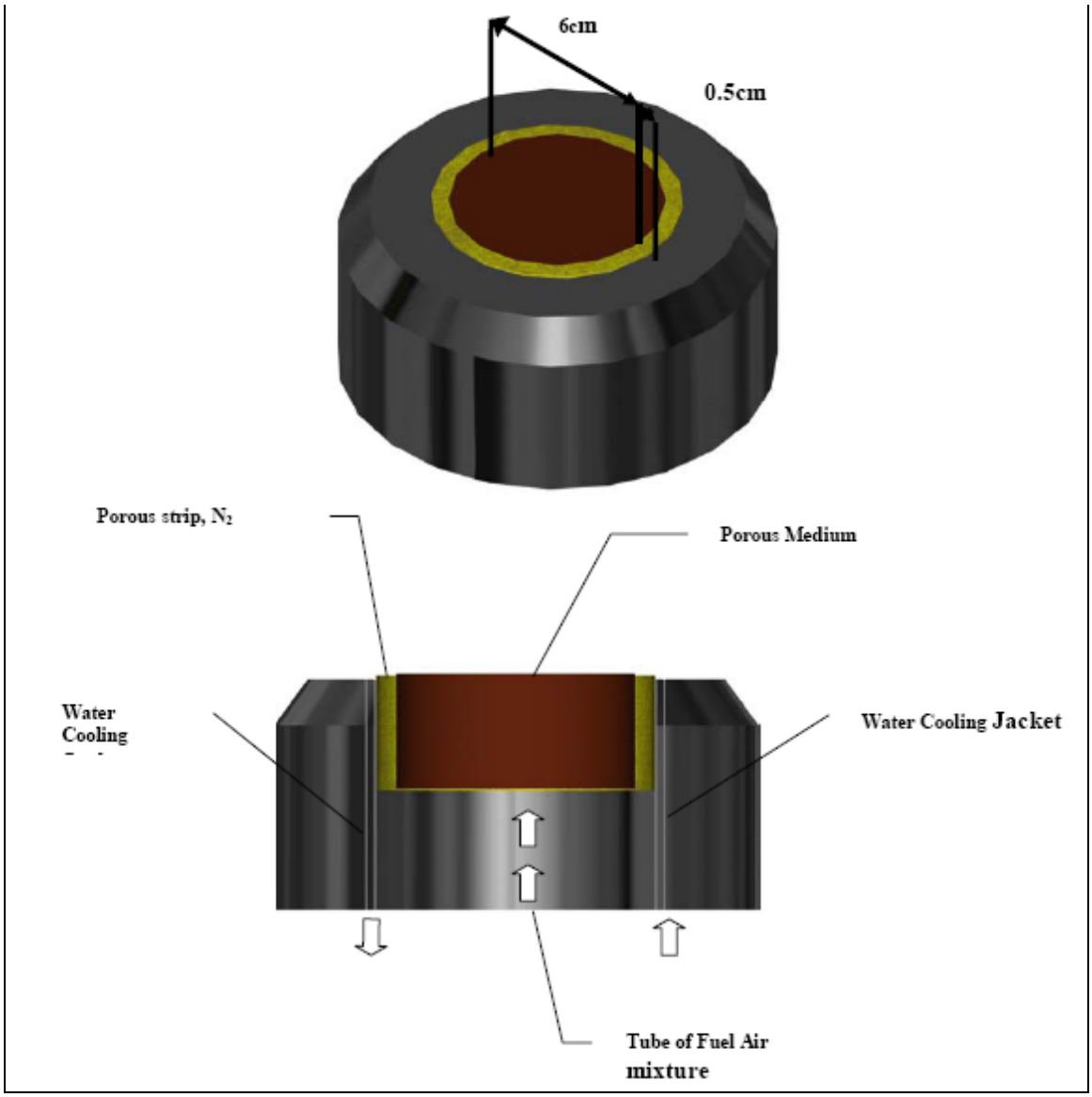

Figure 2.8: Flat flame burner system

III. Results and Discussions

3.1 Extinction Limits

The flame extinction limits of pure CH_4 [99.97%] was measured for the qualification and validation of the present twin flame counter flow burner system. **Figure 3.1** shows the extinction values (in %f) of CH_4 at different stretch conditions. The equation **%f = 0.003 K+ 4.6596** represent the linear fit of the measured data with a R^2 value of 0.98.

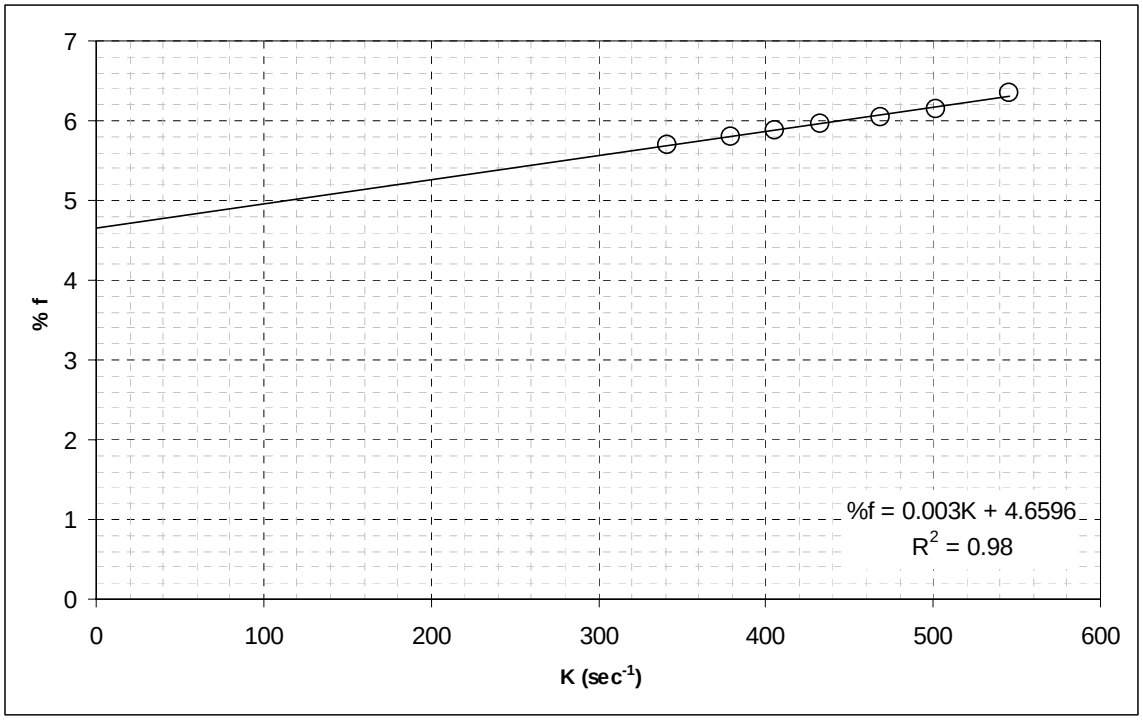

Figure 3.1: Flame extinction limit for pure-methane fuel at different stretch conditions

The zero intercept value of **4.6596** is the extinction value of CH_4 at the zero stretch condition. The estimated repeatability of the measured data is within the **2.5%** of the

mean value (at 95% confidence interval). The measurement uncertainty includes biased error and precision error related to volumetric flow rate measurements (estimated using student-t distribution at 95% confidence interval). Including all the error estimations and rounding off to the significant digits the zero stretch extinction limit (in %f) of CH_4 at the standard laboratory condition is **4.66±0.12**. **Table 3.1** shows the comparison of the present measurements with various extinction limits reported in the literature. The measured value is within 1% of the value reported by Ishizuka and Law, 1982 [**7**] using a similar burner system.

Author	**Method**	**Flame Extinction Limit**
		% Methane
Zabetakis	Propagating flame (tube)	5.00
Andrews and Bradley	Propagating flame (vessel)	4.50
Egerton and Thabet	Flat flame	5.10
Sorenson, Savage and Strehlow	Tent flame	4.00
Yamaoka and Tsuji	Double flame	4.70
Ishizuka and Law	Binary flame	4.80
Present Investigation	Twin flame	4.66

Table 3.1: Extinction limits for pure-methane and pure-propane fuels [Source: Refs. **7** and **18**]

Two assumptions are made to calculate the zero-stretch extinction limit of the CH_4 flames: (i) the global stretch rate represents the local stretch rate, and (ii) the extinction limit and the stretch rate vary linearly toward the zero-stretch limit. Several previous investigations have shown the validity of the first assumption **[18].** Using LDV measurements Kobayashi and Kitano, 1991 [**19**] have shown that the local stretch rate varies proportional to the global stretch rate. Although some recent studies [**10-11**] have

suggested that there may a non-linear relation between the extinction limit, it is estimated that the magnitude error due to the nonlinearity is within the uncertainty of the present measurement. Additionally, **Figure 3.2** shows the CH_4 extinction data from [18] plotted with the data measured in the present investigation. Together they encompass a stretch rate ranging from 30 s^{-1} to 550 s^{-1}. The data shows a linear behavior (with an R^2 value of 0.97) throughout a large range of stretch rates which further supports the second assumption.

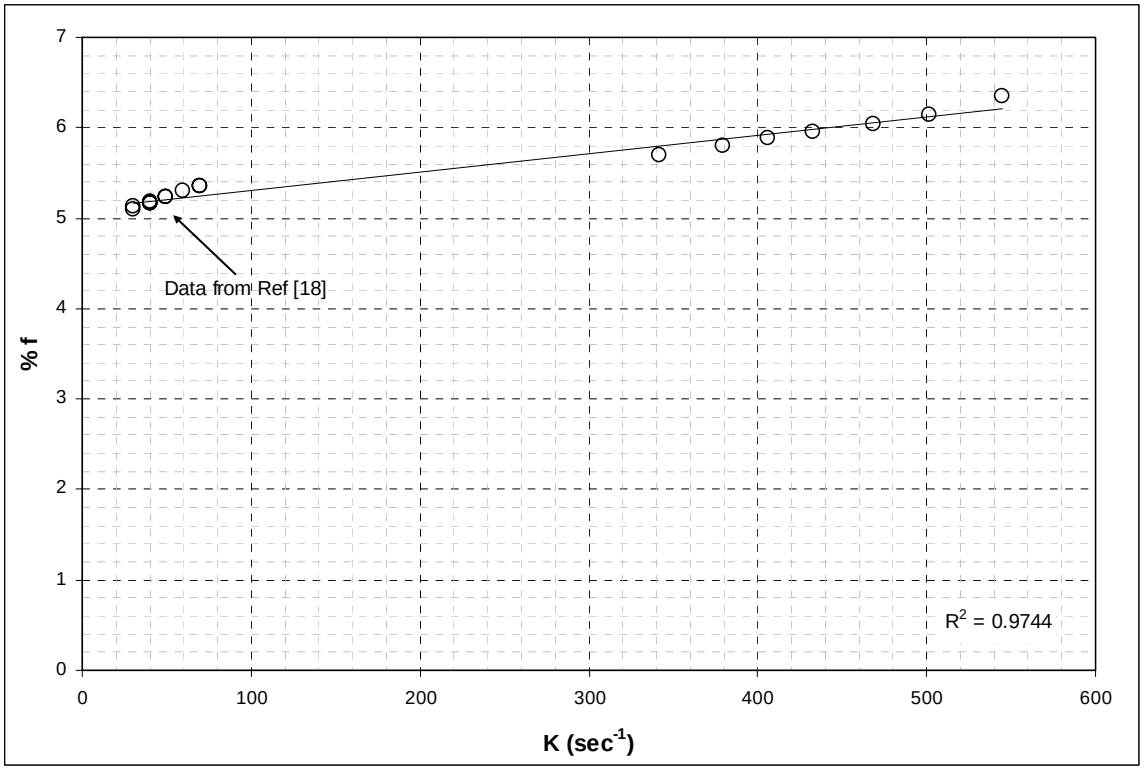

Figure 3.2: CH_4 extinction data from [18] plotted with the data measured in the present investigation

Figure 3.3 shows the extinction of H_2-CO flame at different compositions and stretch condtions. For a given stretch condition flame extinguishes at a much leaner condition with the increase in H_2 concentration in the mixture. This is due to the increase in CO

oxidation rate [$CO+OH \rightarrow CO_2 + H$] and branching [$O + H_2 \rightarrow OH + H$; $OH + H_2 \rightarrow H_2O + H$; $H + O_2 \rightarrow OH + O$] with the increase in H_2 in the mixture. For a given mixture composition, flame extinguishes a much richer condition with the increase in global flame stretch. It is interesting to note that a large change in the extinction behavior of flame occurs with a small addition of H_2 (< 10%) in the mixture. However, with the addition of more H_2 the change is less significant. This is consistence with the sensitivity analysis of H_2-CO kinetics reported by Vagelopoulos and Egolfopoulos, 1994 [**9**]. Initially a small H_2 in the mixture significantly accelerates the mass burning rate due to the fact at this composition regime the $CO+OH \rightarrow CO_2 + H$ primarily control the burning rate. With the further addition of H_2 the hydrogen kinetics kicks-in and the burning rates are less affected by the CO oxidation reactions. **Figures 3.3** shows the normalized sensitivity analysis data reproduced from **Ref [9]** with a modified nomenclature consistence with this report. The analysis clearly shows that the reduction of CO oxidation rate sensitivity on burning rate with the increase in H_2 concentration in the mixture.

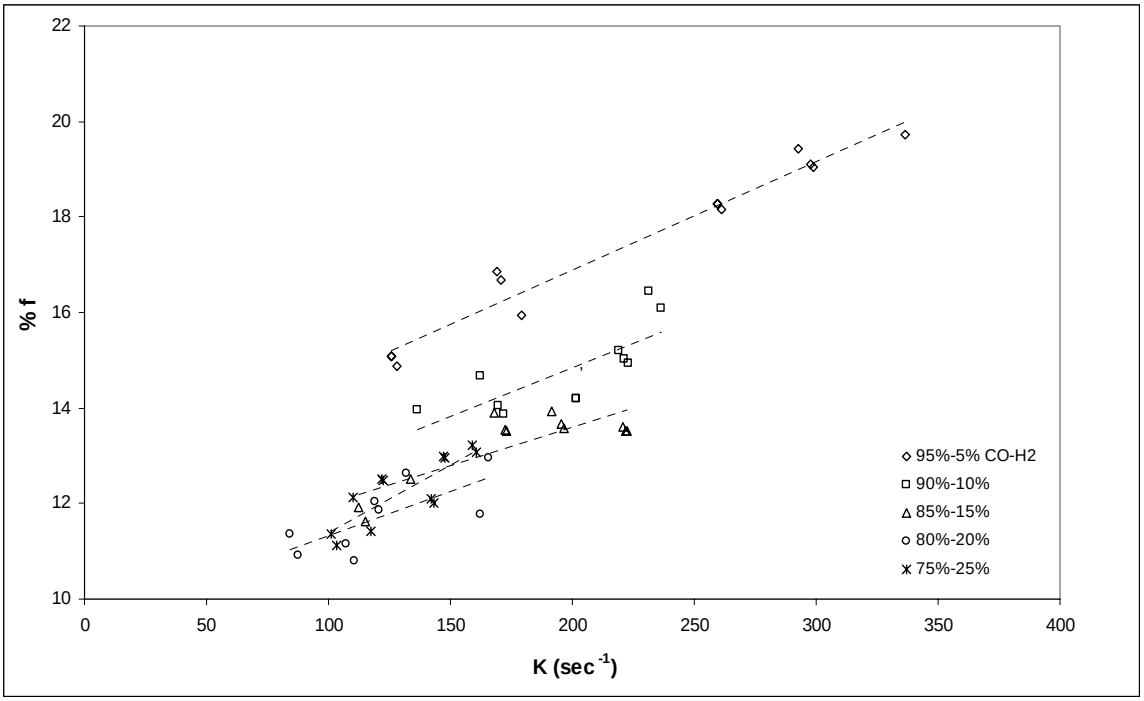

Figure 3.3: Flame extinction limits of H_2 –CO fuel blend composition at different stretch conditions

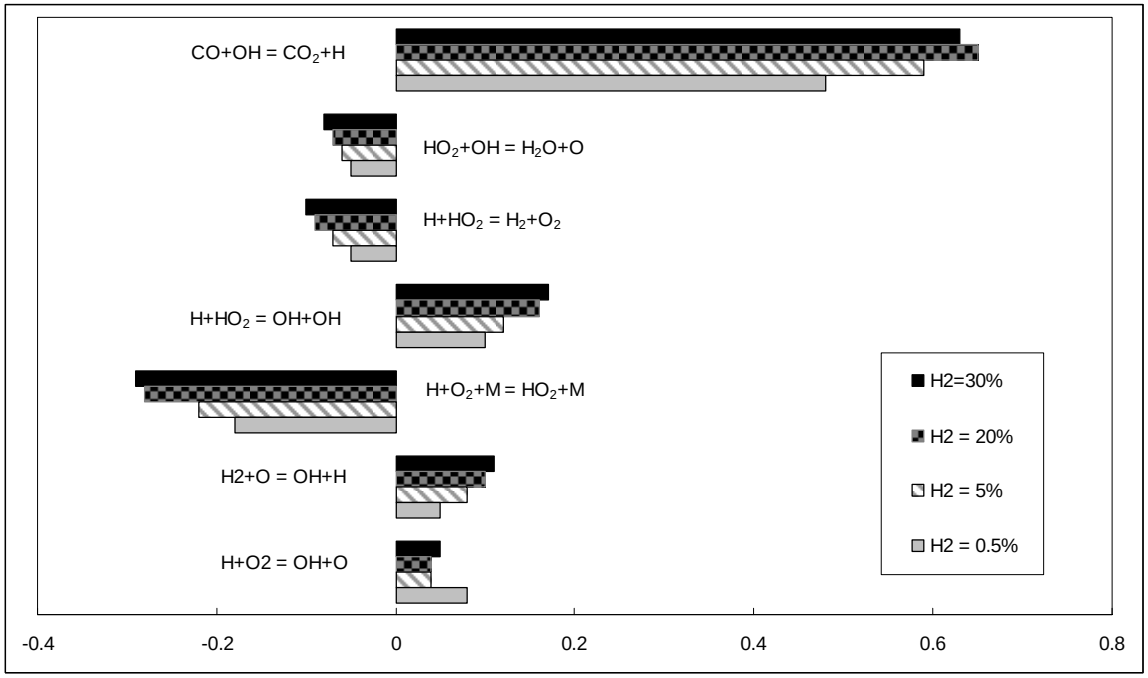

Figure 3.4: Normalized sensitivity analysis data [Reproduced from **Ref 9**]

Figures 3.5 show the variation of zero stretch extinction limits of H_2-CO flames at different H_2 concentration in the fuel mixture. The zero-stretch extinction limit of H_2-CO decreases (from rich to lean) with the increase in H_2 concentration in the mixture. As discussed earlier this due to the branching and increase in the oxidation rate of CO with the increase H_2 concentration in the mixture. The maximum overall uncertainty of the measurement is less than ±2.5% of the mean value. The solid-line in the **Figure 3.5** represents the extinction limits of H_2-CO mixtures calculated using the Le Chatelier's rule. The average difference between the measured data and the Le Chatelier's calculation is around ~ 7%.

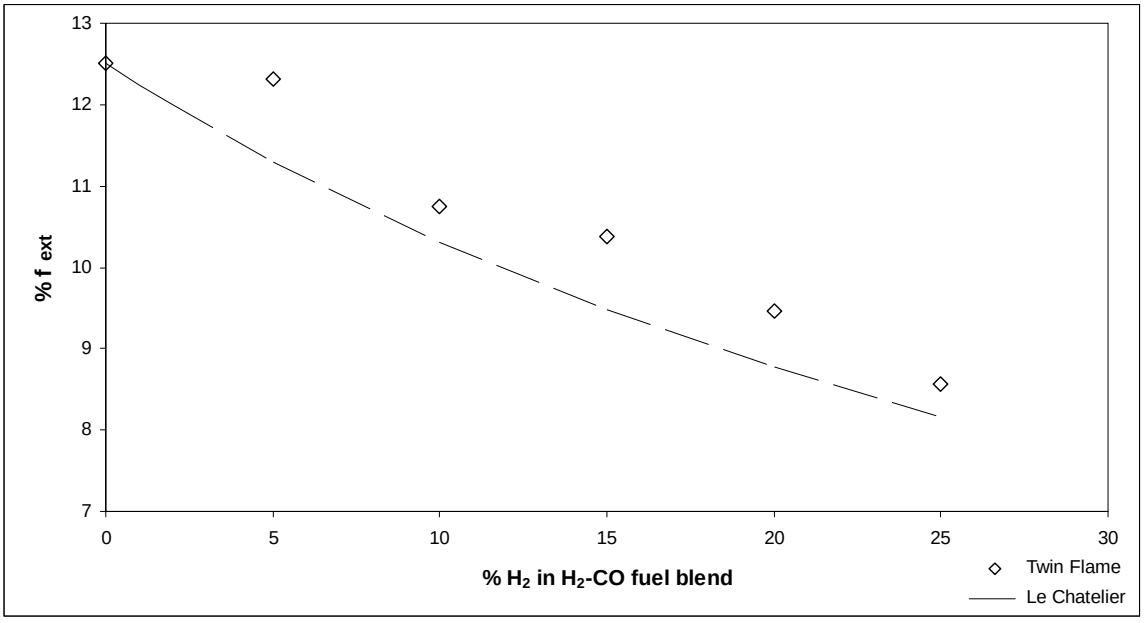

Figure 3.5: Flame extinction limits of H_2-CO fuel blends at different compositions and stretch conditions

In binary or ternary fuel mixtures, the fuel with lower flame speed generally acts as an inhibitor for the flame. For H_2-CO mixtures the CO acts as a flame inhibitor due to its very slower flame speed ($CO+O_2$ does not have any discernible kinetic pathways and requires large activation energy) and thus causes the discrepancies in measured and calculated values. Recently the authors have also reported that [**20**] the discrepancies of measured and calculated values are especially significant (as high as 19%) for hydrocarbon fuel blends where both of the fuel components have comparable burning velocities and fuel properties. The 'inhibitor' effects of fuels with lower flame velocities are especially pronounced for hydrocarbon mixtures with similar laminar burning velocities. It is apparent that the prediction accuracy of Le Chatelier rule is higher for fuel blends with less 'flame inhibitor 'effects.

3.2 Flame Structures

Figure 3.6 shows the measured maximum flame temperature of H_2-CO mixtures as the flame approaches to the extinction limit. It appears that regardless of the mixture compositions the extinction temperature approaches to a common value prior to the extinction. The extinction temperature was found to be **740 (±10) ^{0}C** for the H_2-CO mixtures. It is evident the loss processes overtake the energy release mechanisms at this temperature and extinguishes the flame by ceasing the branching reactions. The measured extinction flame temperature can be effectively used to validate different heat loss models used in various computational schemes.

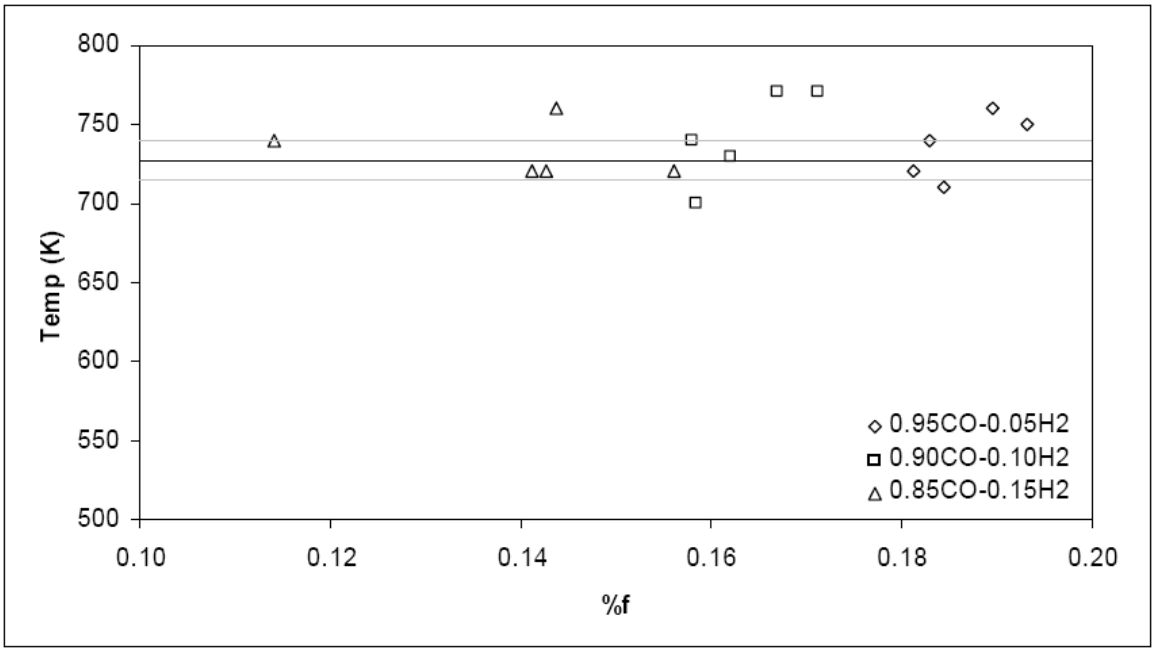

Figure 3.6: Maximum flame temperature of CO-H_2 mixtures as flames approach to extinction

Figures 3.7 and 3.8 show the measured normalized chemiluminescent emissions of OH^* and CH^* (at the maximum temperature location) from H_2-CO flames prior to extinction. The chemiluminescent signals were normalized with the signals acquired from flames subjected to an experimentally attainable maximum stretch rate. The chemiluminescent of OH^* and CH^* radicals can be effectively used to track the changes in flame characteristics [**21**] as it approaches extinction. Additionally these active radicals are prime indicators of the heat releases process. The OH chemiluminescent data indicates that mixtures with more than 5% H_2 content, the OH radical concentration reduces (within the experimental uncertainties) to an extinction value prior to the flame extinction regardless of mixture compositions. In the present chemiluminescent measurement the fuel compositions were kept constant while increasing the global

stretch rate. The measurements were taken at experimentally achievable flame conditions (equivalence ratio) very close to the extinction. Thus the data can be interpreted in the context of a flame condition (experimentally achievable) at which any increase in the stretch rate will cause the flame to extinguish. It is interesting to note that for 5%-95% H_2-CO mixtures the OH^* concentration remains high (in compared to other mixture conditions) even prior to extinction. This is due to the transitional kinetic of H_2-CO at this mixture conditions. The CH^* chemiluminescent emission data does not show any definitive monotonic trend prior to extinction. Prior to extinction the CH^* radical concentration decreases with the increase in H_2 concentration in the mixtures.

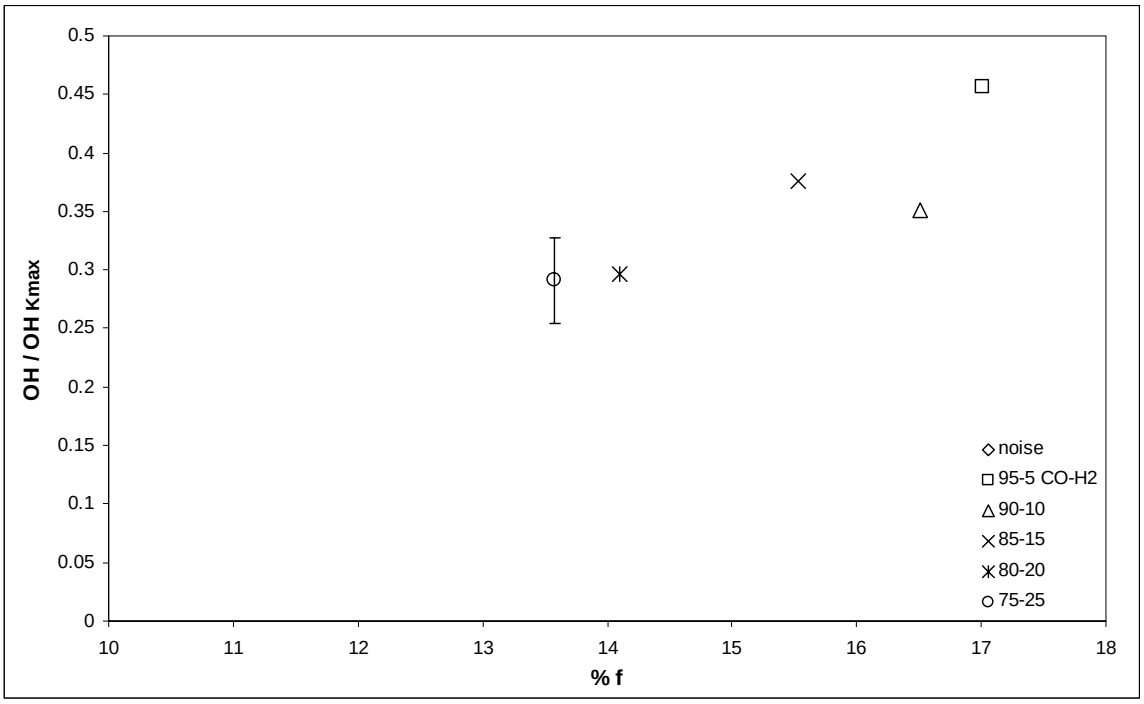

Figure 3.7: Chemi-luminescent emissions of OH^* radicals measured from H_2 –CO fuel blends as flames approach extinction

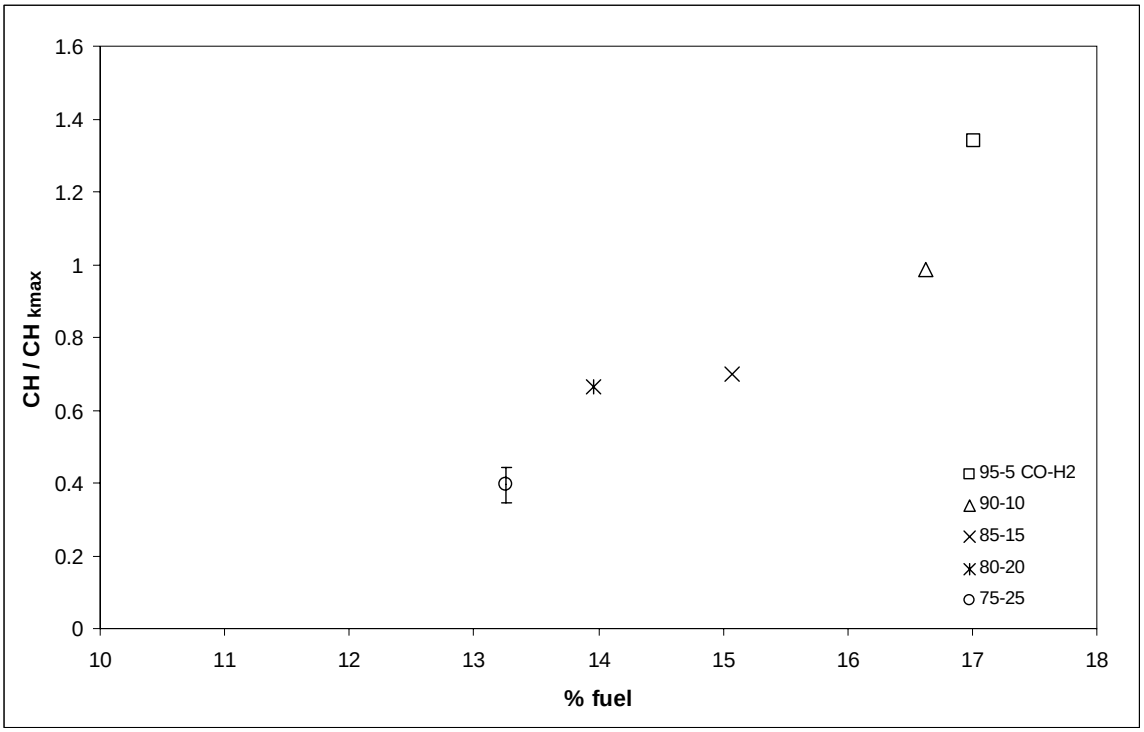

Figure 3.8: Chemi-luminescent emissions of CH^* radicals measured from H_2 –CO fuel blends as flames approach extinction

The OH emission data suggest that as the flames approach extinction condition, production of OH decreases due to the termination of the branching reactions. Although most of the computation studies [**22-23**] showed that they are no kinetically driven limit criterion for flame extinction, in reality extinction occurs when the decrease of OH production causes a lower limit in laminar burning velocity beyond which the loss processes suppress the energy release. However the CH chemiluminescent emissions do not attain the asymptotic value close to the extinction [**Figure 3.8**]. This is due to the fact that laminar burning velocity of a fuel mixture is primary determined by the radicals and atoms such as OH, O, and H [**24-27**]

3.3 Laminar Burning Velocity

The laminar flame velocity at extinction of H_2-CO flames at different concentration of H_2 in the mixtures were measured using the flat flame burner method. The measurements are needed to support the argument that close to the extinction equivalence ratio the laminar burning velocity attains a non-zero asymptotic value. The flat-flame burner is chosen due to the fact that it yields very accurate burning velocity measurements for lean flames. However, the extinction occurs in flat-flame earlier in compared to the twin-flame counter flow burner. Thus extinction conditions were also measured for H_2-CO flames using the flat flame burner at different values of H_2 concentration in the mixture. The extinction conditions in terms of percent fuel in the mixture (%f) are shown in **Figures 3.9.** It is important to note that the extinction values measured in the flat flame configuration are not actually apparatus independent flame extinction values. They are tied with the specific experimental conditions used in the present investigation. The values are reported to couple the flame velocity measurements (in flat flame configurations) with the zero stretch extinction measurements. Clearly, extinction occurs earlier in the flat flame burner in compared to the counter flow configuration. Since the extinction values are at the adiabatic conditions the difference in extinction values underscore the effect of only flame stretch. The flame extinguished earlier due to the termination of chain branching reactions on the curved flame surface. However, it is interesting to note that despite a higher extinction value in the flat flame the relation between the H_2 concentration in the H_2-CO mixtures and the extinction limits of the mixture in both burner configurations show similar trends.

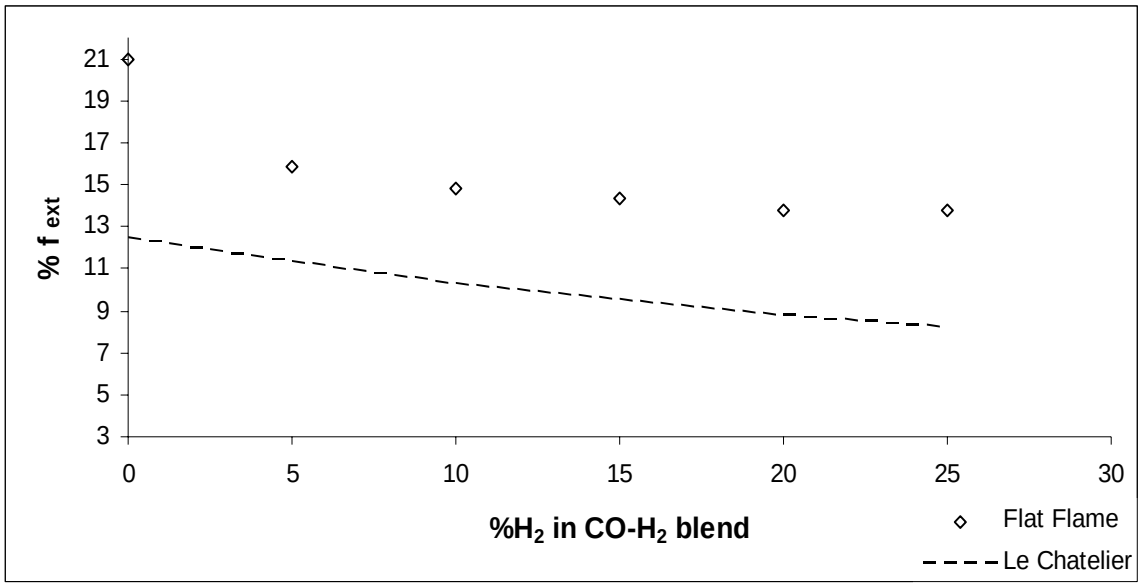

Figure 3.9: Flame extinction limits of H_2-CO fuel blends at different compositions and stretch conditions

The laminar flame velocity at extinction of H_2-CO flames at different concentration of H_2 in the mixtures were measured using the flat flame burner, which are shown in **Figure 3.10**. The flame velocity data clearly indicate that regardless of the fuel types and compositions, the flame ceases to propagate when the laminar flame velocity reduces to a critical value. This supports the fact that despite there exists no extinction flame speed based on the theoretical calculation, one-dimensional premixed flame extinguished when the mixture laminar flame speed falls below a certain threshold. This limiting flame speed is tied with the loss mechanisms and only weakly affected by the rate parameters. The average laminar flame velocity at extinction for H_2-CO measured in the present flat flame burner configuration is 3.77(±0.38) cm/s.

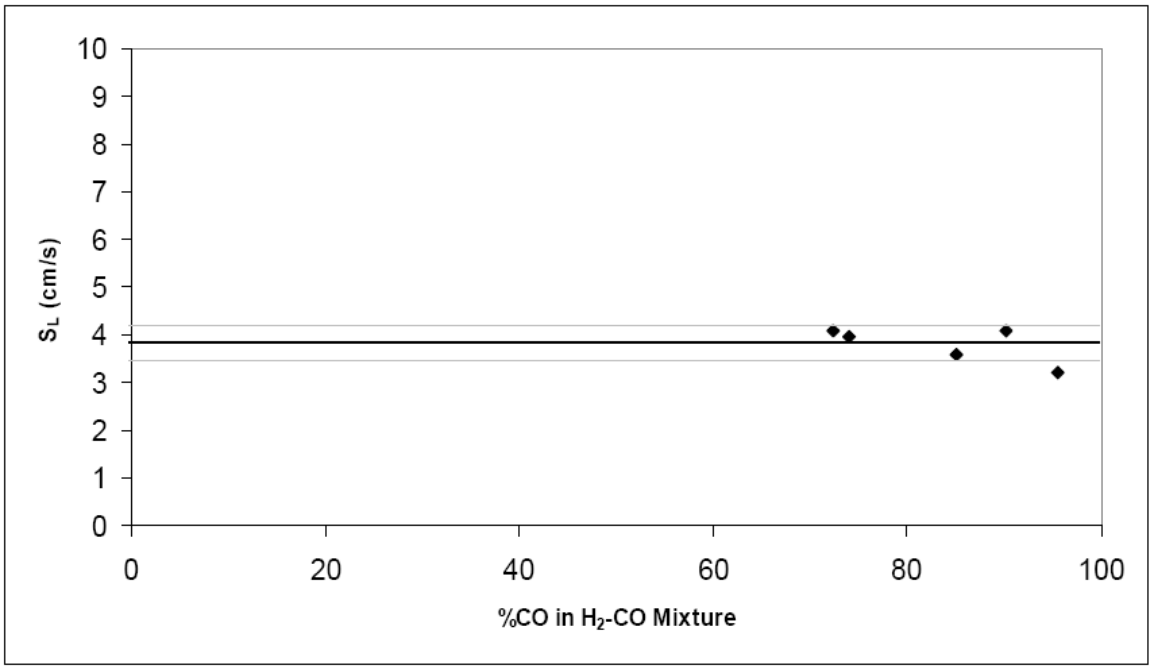

Figure 3.10: Average laminar flame velocity of 3.77 (±0.38) cm/s measured close to extinction for H_2 - CO flames, measurements obtained using Flat flame burner

The observation is further confirmed by the measured flame temperatures at different fuel compositions [**Figures 3.11**]. It was found that close to the extinction the flame temperature of different H_2-CO mixtures collapses to a single value (within the experimental uncertainties). The flame temperatures at extinction (close to the extinction) for different compositions H_2-CO mixture measured in the flat flame burner configuration is 845 (±32)^{0}C.

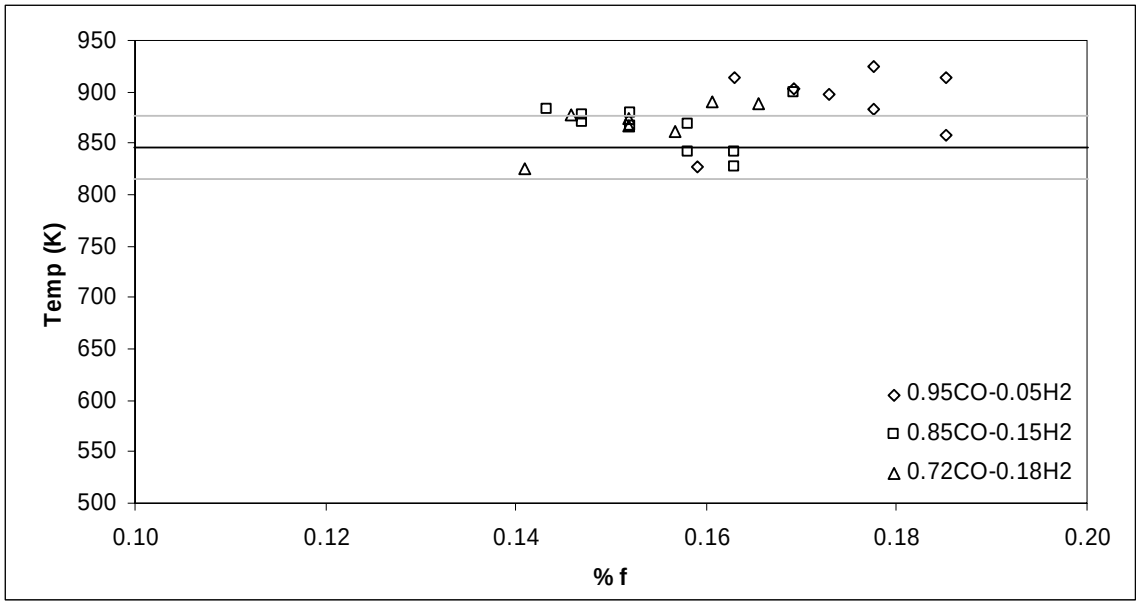

Figure 3.11: Flame temperature of 845(±32)°C measured close to extinction for various H_2-CO fuel blend compositions using Flat flame burner

As indicated earlier several classical theories [**28**] have described a direct correlation between the laminar flame velocity and the concentration of intermediate radicals and atoms such as OH, H and O. These theories postulate that the rate of diffusion of active radicals into the unburned gas region determine the flame speed of particular fuel mixtures. These theories are particularly helpful to compute the laminar flame speed of fuel mixtures since simple additive formulas are not adequate to predict the laminar flame speed of fuel blends [**24-27**]. In the present study qualitative planar laser induced fluorescence images of OH radical concentration in a one-dimensional flame were captured for different compositions of H_2-CO mixture. **Figure 3.12** shows the OH radical concentration in a 24%-76% H_2-CO flame as it approaches the extinction point. The OH radical concentration decreases significantly as the flame approaches the extinction

value. In addition it was also observed that close to the extinction point, flames of different compositions of H_2-CO have similar distribution of OH radicals.

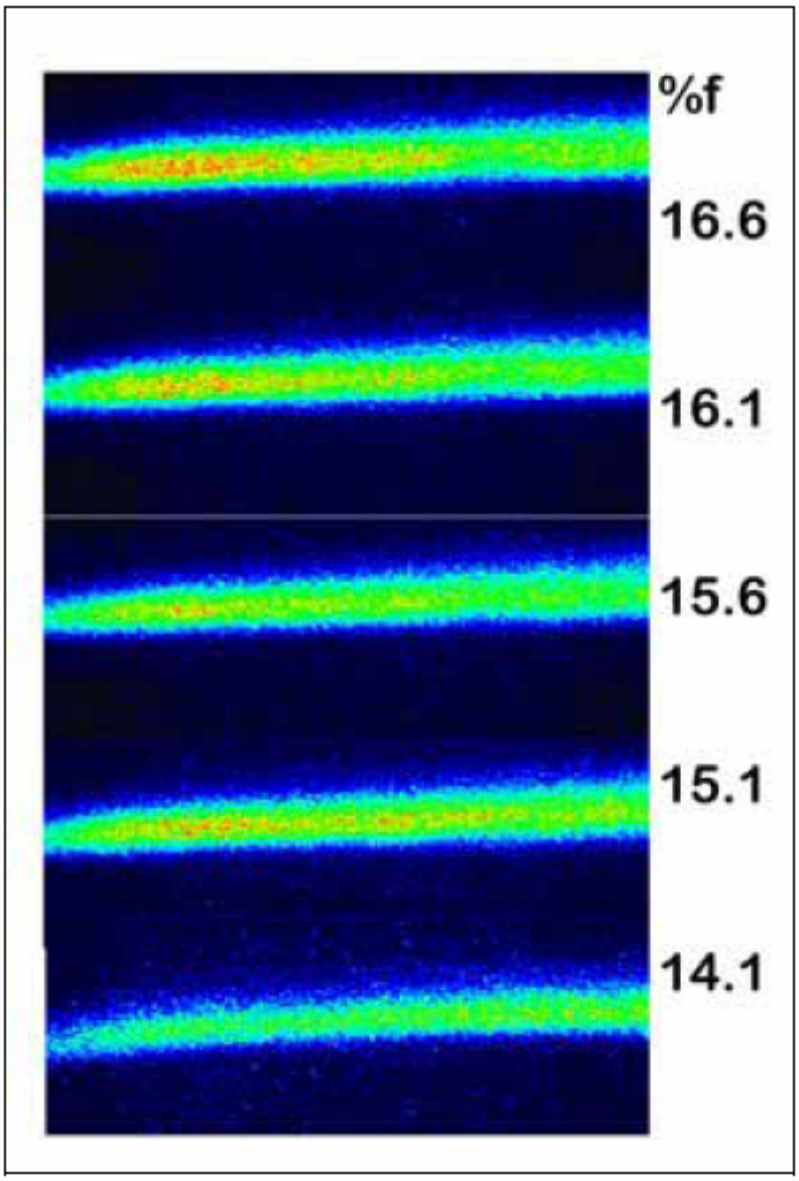

Figure 3.12 : OH radical concentrations measured close to extinction in 24% H_2 – 76% CO fuel blend compositions for various %f, as measured using Flat flame burner

3.4 Flame Extinction in an Oscillatory Strained Field

Figures 3.13 and 3.14 show the digital images of H_2-CO flames distorted by an externally excited oscillatory [300 Hz and 500 Hz] flow-filed. An externally perturbed H_2-CO twin flame was not experimentally achievable for the mixture conditions used in the present investigation. The figures also show the H_2-CH_4 subjected to same oscillatory strained field. With the introduction of an acoustic perturbation the bottom half of the twin-flame becomes unstable quickly and oscillates with the same frequency of the external excitation. At this point a slightest perturbation in the flow-field causes the flame to extinguish. Currently more experiments are ongoing to locate a stable flame regime with externally imposed flow perturbations.

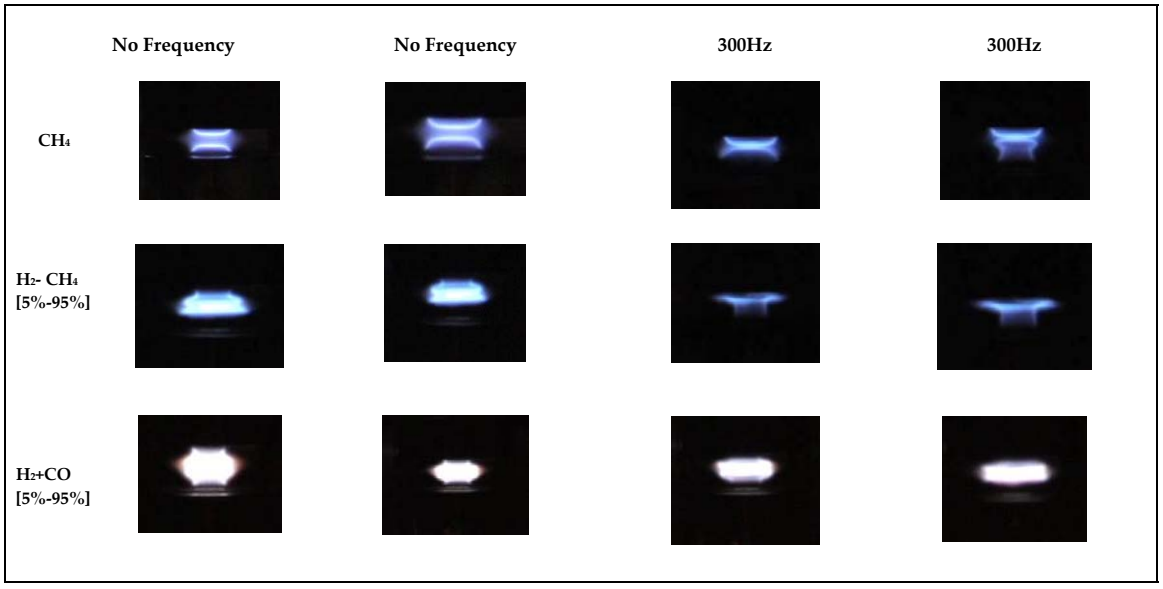

Figure 3.13: H2-CO twin-flames distorted with an acoustic oscillation of 300Hz

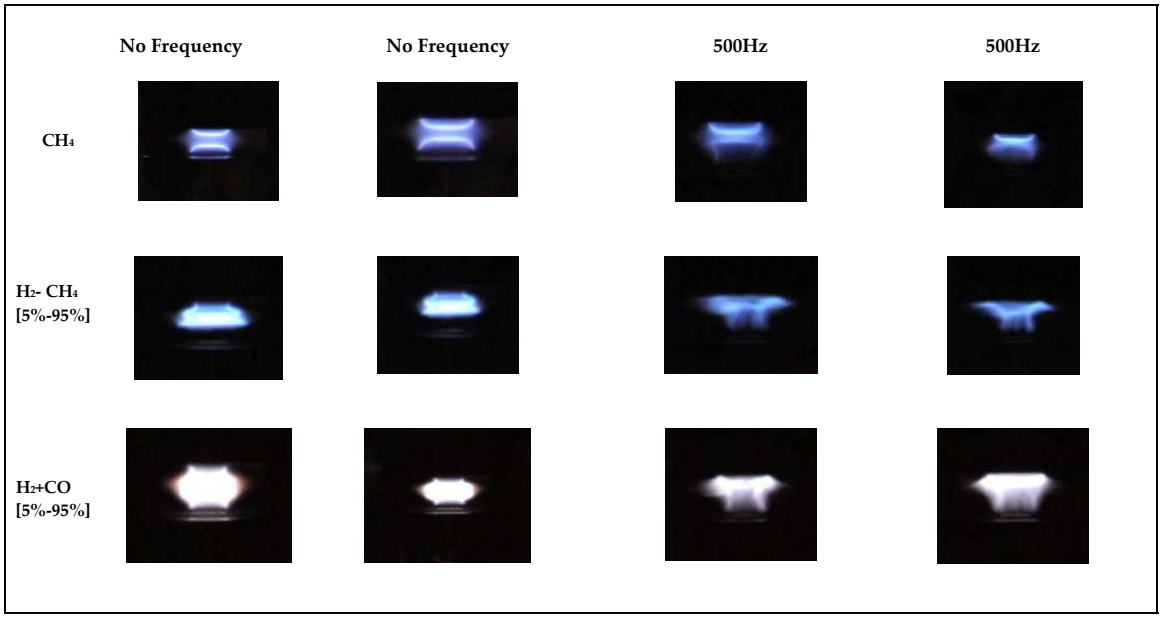

Figure 3.14: H2-CO twin-flames distorted with an acoustic oscillation of 500Hz

Conclusions

Despite their practical importance, the fundamental flame data of hydrogen fuel blends are scarce. These data such as flame extinction limits or critical velocity gradients are necessary not only to aid the development efforts of fuel flexible combustors but also to provide data for the verification and validation of computational schemes. The report presents new data on flame extinction measurements of H_2-CO premixed flames at different mixture compositions. The study concludes that:

1. The flame extinction limits of H_2-CO fuel blends decreases with the increase in H_2 concentration in the mixture.
2. The average difference between the measured extinction limits and the Le Chatelier's calculation is around ~ 7%.
3. Premixed flames of H_2-CO extinguish when the laminar flame velocity falls below a critical value. In the flat flame burner configuration, the critical laminar flame velocity at extinction for H_2-CO measured is 3.77(±0.38) cm/s.
4. Regardless of the mixture compositions the extinction temperature approaches to a common value prior to the extinction. The extinction temperature was found to be 740 (±10) ^{0}C for H_2-CO mixtures.
5. The inflame concentration of OH radical decreases as the premixed flame of fuel blends approaches the extinction equivalence ratio.

6. An externally perturbed H_2-CO twin flame was not experimentally achievable for the mixture conditions used in the present investigation.

Acknowledgement

The initial burner quantification [extinction limits of pure methane] was performed under a previous DOE award DE-FG26-03NT41917 (Project Manager Ben Hsieh). The author likes to thank Professor S. R. Gollahalli of the University of Oklahoma for providing the Flat Flame Burner Facility for the present study. The authors like to acknowledge the help of Combustion and Propulsion Laboratory research assistants Rogelio Franco, Deepthi Davu, Sharmin Nawsher, Mahbub Ahmed Chendhil Periasamy, and Norman Love during experiments

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